SLAC-PUB-13945

# Tunneling density of states as a function of thickness in superconductor/ strong ferromagnet bilayers

S. Reymond,[1] P. SanGiorgio,[2,*] M.R. Beasley,[1] J. Kim,[3] T. Kim,[3] and K. Char[3]
[1]*Department of Applied Physics, Stanford University, Stanford, CA 94305*
[2]*Department of Physics, Stanford University, Stanford, CA 94305*
[3]*Center for Strongly Correlated Materials Research, School of Physics, Seoul National University, Seoul 151-742, KOREA*
(Dated: February 2, 2008)

We have made an experimental study of the tunneling density of states (DOS) in strong ferromagnetic thin films (CoFe) in proximity with a thick superconducting film (Nb) as a function of $d_F$, the ferromagnetic thickness. Remarkably, we find that as $d_F$ increases, the superconducting DOS exhibits a scaling behavior in which the deviations from the normal-state conductance have a universal shape that decreases exponentially in amplitude with characteristic length $d^* \approx 0.4$ nm. We do not see oscillations in the DOS as a function of $d_F$, as expected from predictions based on the Usadel equations, although an oscillation in $T_c(d_F)$ has been seen in the same materials.

One of the incompletely solved problems in conventional (noncuprate) superconductivity is the interaction between superconductivity and magnetism. This issue arises most prominently in the context of the so-called magnet superconductors (e.g., $CeCoIn_5$) and in the superconductor/ferromagnet (SF) proximity effect. One striking effect expected for a superconductor in the presence of an exchange field is the existence of spatial modulations of the superconducting pair wave function (see for instance Ref. [1]). These oscillations occur in a new superconducting state where the center of mass of pairs acquires a non-zero momentum. This state was predicted 40 years ago and is known as the Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) state [2, 3]. In the case of the SF proximity effect, this oscillating pair wave function is expected to exponentially decay into the F layer. These oscillations, in turn, are predicted to lead to oscillations in the critical temperature of SF bilayers [4, 5], inversions of the DOS [6, 7], and changes in the sign of the Josephson coupling in SFS sandwiches [8] (creating a so called $\pi$-junction), as the F layer thickness, $d_F$, is varied. Even more exotic predictions include possible odd-frequency triplet superconductivity [9].

Indeed, a vast theoretical literature now exists regarding the SF proximity effect, mostly concerning systems with a uniform magnetization in the F layer in the dirty quasiclassical limit (i.e. where all the characteristic lengths are larger than both $\lambda_F$, the Fermi wavelength, and $\ell$, the mean free path). In this situation, the superconducting properties can be calculated using the Usadel equation [10]. Still, explicit predictions may differ depending on the importance of scattering processes [1, 11] or on the boundary conditions, which can be resistive [5] or magnetically active [12].

Experimentally, phase sensitive measurements in some SFS structures clearly demonstrate the existence of $\pi$-junctions [13, 14, 15]. On the other hand, critical temperature, $T_c$, measurements in SF structures have shown a variety of behaviors as a function of $d_F$. Everything from monotonic dependence to step-like features, small dips and oscillations have been reported [5, 16, 17, 18, 19, 20]. It has been pointed out that important parameters such as the size of the exchange field, $E_{ex}$, and the boundary resistance, $\gamma_B$, can evolve naturally as a function of $d_F$ [21]. What makes these results particulary hard to interpret is that all of these changes typically take place within a few nanometers, just like the expected oscillation of the superconducting wave function inside the F material.

In short, the situation is complicated both theoretically and experimentally, and no clear, comprehensive understanding has emerged. This suggests that some new experimental approaches that probe superconductivity directly inside the F material might be helpful.

In this paper we present density of state spectroscopy studies using tunneling junction located on the F side of $Nb/Co_{0.6}Fe_{0.4}$ bilayers. CoFe is a strong ferromagnet with a Curie temperature of approximately 1100 K widely used in magnetic tunnel junction devices due to the high quality of the interface it makes with aluminum oxide, which is now the standard choice for tunneling barriers. The critical temperatures of similarly deposited Nb/CoFe bilayers, but with a thinner Nb layer (18 nm), show an oscillatory behavior as a function of $d_F$: a slight dip is noticeable before saturation at large $d_F$. A quantitative analysis of these data based on the Usadel equations appears elsewhere [22]. Based on these data, we expect that any inversion in the DOS that may arise in our samples would occur at the same thickness as the dip in $T_c$, which is between 1 and 2 nm. Thus, we performed tunneling spectroscopy on samples with thicknesses ranging from 0 to 4.5 nm in increments of 0.5 nm.

Our results are remarkable in that we find that the deviations of the density of states from that of the normal state exhibit a precise scaling in amplitude over many orders of magnitude as $d_F$ is increased. No oscillations in the sign of these deviations are observed.

Our junctions are deposited and patterned entirely *in*

SIMES, SLAC National Accelerator Center, 2575 Sand Hill Road, Menlo Park, CA 94309
Work supported in part by US Department of Energy contract DE-AC02-76SF00515.

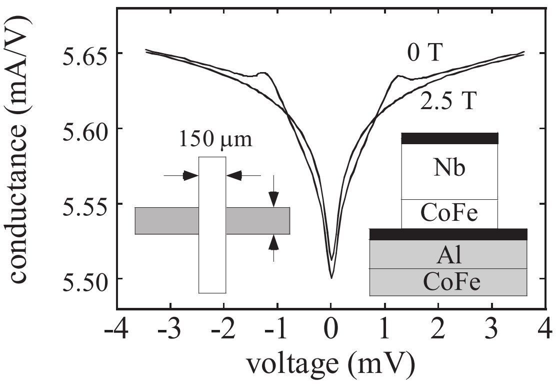


FIG. 1: Raw conductance data taken at 0.5 K in a Nb/CoFe/AlO$_x$/Al/CoFe junction at zero field and above the Nb critical field. Here $d_F = 2.5$ nm. Left inset: sample top view. Right inset: junction cross section. The black areas represent Al oxide layers.

*situ* in a DC magnetron sputtering system, described here [22]. The full geometry of the tunneling structures is pictured in the inset of Fig. 1. A thin (3 nm) CoFe layer which suppresses superconductivity in the Al electrode is deposited first — we see no superconducting transition in the Al above 0.3 K — then an atomic oxygen source is used to fully oxidize the barrier to a thickness of ≈ 2 nm. The stencil mask is then changed and without breaking vacuum a CoFe layer and then a Nb layer are immediately deposited, followed by a very thin Al cap to protect the Nb layer from oxidation. In order to establish a robust superconducting state in the S layer, we deposit approximately 50 nm of Nb, which is thick enough that there should be only small changes in $T_c(d_F)$ [30]. Typical junction resistances are roughly 200 Ω, with a trend to higher junction resistances with increased CoFe thickness.

Measurements shown here are taken at 0.5 K using standard lock-in techniques to measure the differential resistance; variations as small as few parts in 10,000 could be distinguished. This resolution is crucial since when $d_F$ exceeds 1 nm the superconducting signal becomes very weak, reaching less than a part per thousand at $d_F = 3$ nm.

Figure 1 shows typical conductance spectra taken at 0.5 K in zero-field and with a perpendicular field of 2.5 T, just above the critical field, $H_{c2}$. The zero-field curve clearly reveals the superconducting density of states, but it is superposed on top of a zero-bias anomaly. The high field data show the zero-bias anomaly on its own. This anomaly is a V-shaped curve, centered at zero volts, with the conductance at 1 mV typically two percent larger than the zero-bias conductance. The main experimental difficulty lies in extracting the superconducting DOS from this zero-bias anomaly. To do this, we divide the

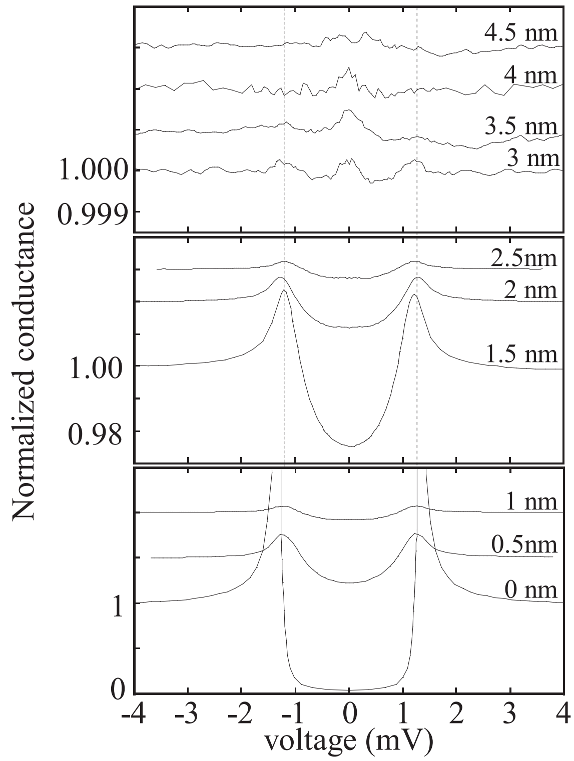


FIG. 2: Normalized conductances taken at 0.5 K for various CoFe thickness indicated inside. From the bottom to the top plot the vertical scale is successively amplified. The curves are shifted for clarity. The dashed line shows that the peaks maximum remains unchanged from 0.5 to 3 nm.

zero-field curve by the in-field curve. This straightforward procedure alone was sufficient for the thicker barriers, because no change in the normal conductance was seen between perpendicular and parallel applied fields or as a function of field strength. However for samples with thin CoFe layers (less than 1.5 nm), an additional zero-bias resistance peak increasing with the applied field was seen above $H_{c2}$, similar to previous studies [23]. For these samples, the zero-field normal-state background could be calculated by studying the field-dependence of this feature and extrapolating it to zero-field.

Figure 2 shows the resulting curves for all measured thicknesses. Note that in the top panel the scale is amplified about a thousand times. Beginning at the bottom of the figure, we see that for $d_F = 0$ a clean DOS curve for Nb is obtained, as expected. A BCS fit to this curve gives 1.3 meV for the gap energy. When the thinnest (0.5 nm) F layer is added to the superconductor, the tunneling spectrum is abruptly altered from a BCS-like shape to a much more smeared shape with substantial conductance below the gap energy. As $d_F$ increases further, the superconducting features in the tunneling conductance are strongly attenuated. Above 2.0 nm, a narrow zero-bias conductance peak develops which is suppressed in fields

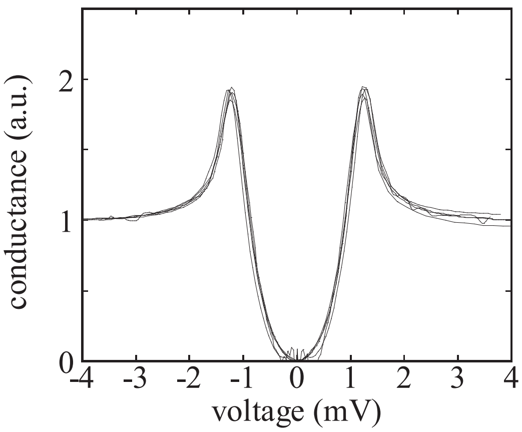


FIG. 3: Superposition of five scaled conductance curves for $d_F = 0.5$ to 2.5 nm. The $d_F = 2.5$ nm curve is scaled by a factor of 500.

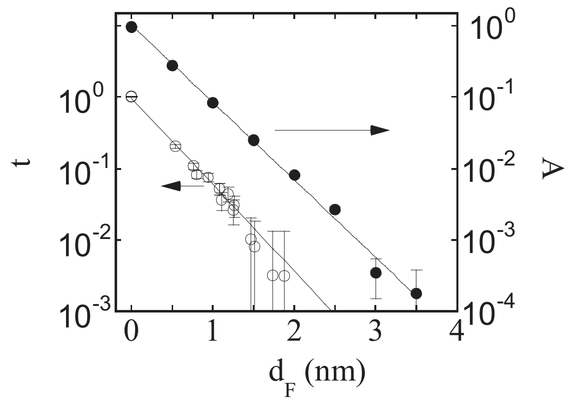


FIG. 4: Scaling factor, $A(d_F)$, and normalized transition temperature, $t = (T_c - Min[T_c])/T_c(d_F = 0)$, as a function of $d_F$ for SF bilayers. For $t(d_F)$, only the initial decay is plotted ($d_F < 2$ nm). For both sets of data, the characteristic decay length is 0.4 nm.

greater than $H_{c2}$; this suggests it is related to superconductivity, but due to its weak signal-to-noise ratio, we do not focus on it in the present discussion. When $d_F$ exceeds 3.5 nm, all recognizable features disappear and the normalized conductance is equal to $1 \pm 10^{-4}$.

The most striking observation, though, is that between 0.5 nm and 2.5 nm the spectra can be rescaled onto a single curve. That is to say,

$$\sigma(V, d_F) - 1 = A(d_F)(N(V) - 1) \tag{1}$$

where $N(V)$ is a generic, thickness independent function and $A(d_F)$ is a scaling coefficient defined so that $N(0) = 0$. The $N(V)$ curves derived from the data using this scaling procedure are shown in Fig. 3. The overlap of the curves demonstrates the generic nature of $N(V)$. $A(d_F)$ is plotted in Fig. 4 (full circles). The straight line is an exponential fit with decay length $d^* = 0.4$ nm. Note that for $d_F > 2$ nm, we disregard the narrow peak at $V = 0$ when scaling the curves. The fact that $A(d_F)$ extrapolates to 1 as $d_F$ tends to zero is not required by the scaling procedure. It has the consequence that $N(V)$ can be interpreted physically as the normalized tunneling DOS when some minimal thickness of F material has been deposited below the S layer. Note also that the zero-bias conductance is simply $1 - A(d_F)$ and therefore rises exponentially from zero as $d_F$ increases.

An important characteristic of the generic DOS $N(V)$ is that the sum rule on the DOS is satisfied within one percent. This not only justifies the procedure used to isolate the superconducting DOS, even when it is much weaker than the "normal" background, it also suggests that our tunnel barrier is not weakend by the addition of the CoFe layer. Such weakening would result in an excess current within the gap, raising the total spectral area above unity [24].

It is interesting to compare these DOS data with the $T_c(d_F)$ data reported earlier on related samples with a thinner Nb layer, mentioned above. There are no evident oscillations in $A(d_F)$ near $d_F \approx 2$ nm, where an oscillation is seen in $T_c(d_F)$. On the other hand, in Fig. 4, we compare the initial drop in $T_c$ (open circles) with the scaling factor, $A(d_F)$, derived from the DOS. Specifically, we plot the normalized critical temperature, $t = (T_c - Min[T_c])/T_c(d_F = 0)$. Remarkably, both sets of data show an exponential decrease with the same characteristic length scale.

The absence of oscillations in our DOS data contrasts with a previous experimental study on a different material [25]. In that study, a weak ferromagnetic alloy ($Pd_{1-x}Ni_x$) was studied at two different thicknesses and a robust inversion of the DOS was seen. On the other hand, a subsequent study in which the Ni composition (and hence $E_{ex}$) was varied at fixed $d_F$ showed scaling in the DOS similar to that reported here [26]. It is natural that these different approaches should yield similar results, as the relevant magnetic length, $\xi_F$, is given by $\sqrt{\hbar D/2E_{ex}}$, where $D$ is the diffusion constant in the F layer. In addition, the authors of Ref. [26] note that for relatively thick ($d_F \gg \xi_F$) samples such as theirs, scaling of the density of states with exponentially decaying oscillations is predicted by the linearized Usadel equations appropriate to that limit. This is consistent with their data [27], but it is clearly inconsistent with ours.

Almost any application of the Usadel formalism that starts with a large exchange field will predict decaying oscillations in the DOS. In order to prevent these oscillations within this framework, one must include a generic Abrikosov-Gorkov spin-breaking parameter, $\Gamma_{AG} \gtrsim E_{ex}$. Further, to obtain strictly exponential scaling of the DOS (in the $d_F \gg \xi_F$ limit) one must take $\Gamma_{AG}$ to be much greater than $E_{ex}$. In this model, $d^* = \sqrt{\hbar D/4\Gamma_{AG}}$, which would neccessitate $\Gamma_{AG} = 300$ meV to account for our data.

Although the above model gives a qualitative explanation for the two key features of our data (the exponential nature of $A(d_F)$ and the invariance of $N(V)$), it is not clear how accurately it corresponds to the actual physical situation. Our thinnest sample, with $d_F = 0.5$ nm, for instance, is certainly not in the semi-infinite limit, $d_F \gg d^*$. Thus, we would expect finite-size effects, such as a decrease of the gap width, which would prevent it from following the predicted scaling relationship. Further, one must wonder about the origin of $\Gamma_{AG}$ and the seeming unimportance of $E_{ex}$. Previous studies on thin CoFe films have confirmed that near-bulk ferromagnetic ordering exists in films as thin as 1 nm [28], and further, it is reasonable to suggest that given the constant shape of the DOS, none of the important physical parameters are changing significantly over the thicknesses we examine. Therefore, we conclude that a constant exchange field is present in all of our samples with a CoFe layer. Another possible mechanism for washing out the expected oscillations could be lateral variations in $d_F$ [29], but we note that these variations would have to be vary large ($\delta d_F \sim \xi_F$), which seems unlikely. Finally, we note that the Usadel equation is only strictly valid in the diffusive limit, wherein all relevant length scales are larger than the mean free path. From resistivity data, we estimate $\ell = 1.0$ nm, which is somewhat larger than $d^* = 0.4$ nm, which is comparable in size to $\lambda_F = 0.3$ nm. In light of the totality of the considerations above, we come to the position that no reasonable application of the conventional Usadel theory or purely materials problem can account for our results.

In summary, we have measured the tunneling DOS on the F side of SF bilayers and have found a sharp change in the shape of the conductance curves between $d_F = 0$ and $d_F = 0.5$ nm and that for all $d_F > 0$, the shape of the conductance curves is universal. The only dependence on $d_F$ is given by an exponential decrease in the magnitude of the superconducting signal with characteristic length $d^* = 0.4$ nm. We also note a similar exponential decrease in $T_c(d_F)$ in related samples. Finally, we note that we have been unable to reconcile our results with the conventional Usadel equation; thus, new theoretical considerations appear necessary.

We thank L. Litvak and Y. Bazaliy for stimulating discussions. We also thank T. Kontos and O. Valls for helpful communications. The authors acknowledge the support of U.S. DOE, NSF, Swiss National Science Foundation, and KOSEF through CSCMR.

* Electronic address: psangior@stanford.edu

[1] E. A. Demler, G. B. Arnold, and M. R. Beasley, Phys. Rev. B **55**, 15174 (1997).
[2] P. Fulde and R. A. Ferrel, Phys. Rev. **135**, A550 (1964).
[3] A. Larkin and Y. Ovchinnikov, Sov. Phys. JETP **20**, 762 (1965).
[4] Z. Radovic, M. Ledvij, L. Dobrosavljević-Grujić, A. I. Buzdin, and J. R. Clem, Phys. Rev. B **44**, 759 (1991).
[5] Y. V. Fominov, N. M. Chtchelkatchev, and A. A. Golubov, Phys. Rev. B **66**, 14507 (2002).
[6] A. Buzdin, Phys. Rev. B **62**, 11377 (2000).
[7] M. Zareyan, W. Belzig, and Y. V. Nazarov, Phys. Rev. Lett. **86**, 308 (2001).
[8] A. I. Buzdin, L. N. Bulaevskii, and S. V. Panyukov, JETP Lett. **35**, 178 (1982).
[9] F. S. Bergeret, A. F. Volkov, and K. B. Efetov, Phys. Rev. Lett. **86**, 4096 (2001).
[10] K. D. Usadel, Phys. Rev. Lett. **25**, 507 (1970).
[11] L. Crétinon, A. K. Gupta, H. Sellier, F. Lefloch, M. Fauré, A. Buzdin, and H. Courtois (2005), cond-mat/0502050.
[12] H. Doh and H.-Y. Choi (2004), cond-mat/0407149.
[13] V. V. Ryazanov, V. A. Oboznov, A. V. Veretennikov, and A. Y. Rusanov, Phys. Rev. B **65**, 20501(R) (2001).
[14] T. Kontos, M. Aprili, J. Lesueur, F. Genêt, B. Stephanidis, and R. Boursier, Phys. Rev. Lett. **89**, 137007 (2002).
[15] W. Guichard, M. Aprili, O. Bourgeois, T. Kontos, J. Lesueur, and P. Gandit, Phys. Rev. Lett. **90**, 167001 (2003).
[16] C. Strunk, C. Sürgers, U. Paschen, and H. v. Löhneysen, Phys. Rev. B **49**, 4053 (1994).
[17] J. S. Jiang, D. Davidovic, D. H. Reich, and C. L. Chien, Phys. Rev. Lett. **74**, 314 (1995).
[18] T. Mühge, K. Theis-Bröhl, K. Westerholt, H. Zabel, N. N. Garif'yanov, Y. V. Goryunov, I. A. Garifullin, and G. G. Khaliullin, Phys. Rev. B **57**, 5071 (1998).
[19] L. Lazar, K. Westerholt, H. Zabel, L. R. Tagirov, Y. V. Goryunov, N. N. Garif'yanov, and I. A. Garifullin, Phys. Rev. B **61**, 3711 (2000).
[20] O. Bourgeois and R. C. Dynes, Phys. Rev. B **65**, 144503 (2002).
[21] J. Aarts, J. M. E. Geers, E. Brück, A. A. Golubov, and R. Coehoorn, Phys. Rev. B **56**, 2779 (1997).
[22] J. Kim, J. H. Kwon, K. Char, H. Doh, and H.-Y. Choi (2005), cond-mat/0504543.
[23] A. J. Heinrich, J. A. Gupta, C. P. Lutz, and D. M. Eigler, Science **306**, 466 (2004).
[24] G. E. Blonder, M. Tinkham, and T. M. Klapwijk, Phys. Rev. B **25**, 4515 (1982).
[25] T. Kontos, M. Aprili, J. Lesueur, and X. Grison, Phys. Rev. Lett. **86**, 304 (2001).
[26] T. Kontos, M. Aprili, J. Lesueur, X. Grison, and L. Dumoulin, Phys. Rev. Lett. **93**, 137001 (2004).
[27] T. Kontos (2005), private communication.
[28] D. Wang, J. M. Daughton, K. Bussmann, and G. A. Prinz, J. Appl. Phys. **83**, 7034 (1998).
[29] K. Halterman and O. T. Valls, Phys. Rev. B **66**, 224516 (2002).
[30] However the Nb thickness was not tightly controlled and consequently our samples display a non-systematic spread in $T_c$